

Reduced graphene oxide based electrocatalysts for efficient hydrogen peroxide generation

Hyo Won Kim and Bryan D. McCloskey*

Department of Chemical and Biomolecular Engineering, University of California, Berkeley, California 94720, United States
Energy Storage and Distributed Resources Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States

The development of environmental-friendly distributed production of valuable chemicals, including hydrogen, hydrocarbons, ammonia, and hydrogen peroxide, could be enabled by improvements in cost-effective electrochemical technologies. For example, electrochemical oxygen reduction has garnered attention as an emerging alternative to the traditional anthraquinone oxidation process for hydrogen peroxide synthesis. However, the only suitably active electrocatalysts are expensive and therefore limit commercialization of such electrochemical devices. In this talk, we show an extremely selective and efficient graphene-based electrocatalyst to form hydrogen peroxide from oxygen. This electrocatalyst is synthesized by an easily scalable mild thermal reduction of graphene oxide (mrGO) at 100°C and ambient pressure. Few-layered mrGO (*F*-mrGO) is successfully prepared on a porous carbon substrate. We have characterized electrocatalytic selectivity between H₂O and H₂O₂ formation by monitoring O₂ pressure decay in well-stirred, modified electrochemical H-cells with a calibrated, known headspace volume. We also quantify H₂O₂ formation using a standard iodometric titration on the electrolyte after measurements. These techniques provide direct measurements of O₂ consumption and H₂O₂ formation during ORR and are therefore more accurate than traditional rotating ring disk electrode measurements. Interestingly, the synthesized mrGO with few-layered structures shows much higher electrocatalytic activity than aggregated structure. During oxygen reduction, certain variants of the *F*-mrGO electrocatalyst exhibit highly selective and stable peroxide formation activity with nearly no applied overpotential (<10mV) in basic conditions, exceeding the performance of current state-of-the-art precious metal catalysts. Moreover, a combination of spectroscopic structural characterization and *in-situ* Raman spectroelectrochemistry provides strong evidence that sp² hybridized carbon near ether defects, particularly when located at sheet edges, is the most active site for peroxide production. A more detailed discussion will be covered in this presentation.